

**REMARKS**

The Examiner is thanked for the due consideration given the application. This amendment is being filed concurrent with a Request for Continued Examination.

Claims 1, 3-6 and 10-24 are pending in the application. Claims 2 and 7-9 have been canceled. Claim 1 has been amended to better set forth the invention being claimed, and the amendment to claim 1 finds support in the numerous referrals to "PZT" (lead zirconate/titanate) in the specification. Claims 21-24 are new and find support at page 8 of the specification.

No new matter is believed to be added to the application by this amendment.

**Objection to Disclosure**

It is believed that the disclosure is free from informalities.

**Rejection Under 35 USC §112, First Paragraph**

Claims 1, 12 and 13 have been rejected under 35 USC §112, first paragraph, as failing to comply with the written description requirement. This rejection is respectfully traversed.

The compliance with the written description requirement was fully set forth in the amendments filed April 14, 2008 and May 12, 2008. For brevity, these discussions are not repeated here.

Regarding the magnitude of  $a$  in the formula  $Pb_{1-a}RE_bZr_xTi_yTR_zO_3$ , one of ordinary skill would recognize that the

proportion of a must be greater than 0. Otherwise, the ceramic composition would not be a piezoceramic material. Also, claim 1 has been instantly amended so that Pb must necessarily be present in the piezoceramic composition. Further, claim 1 specifically recites a "piezoceramic composition."

The Advisory Action of June 6, 2008 asserts that there is no evidence regarding the proportion of components in the PZT and piezoelectric properties. As evidence thereof, please find attached a portion of Chapter 7 of *Piezoelectric Ceramics* by B. Jaffe, in which the relationship of PZT, phase structure and morphology is discussed. New claims 20-24 have also been presented to better set forth the relation between morphology and piezoelectric properties.

As a result, the claims are in full compliance with the written description requirement.

This rejection is believed to be overcome, and withdrawal thereof is respectfully requested.

**Rejection Under 35 USC §112, Second Paragraph**

Claims 1-20 have been rejected under 35 USC §112, second paragraph, as being indefinite. This rejection is respectfully traversed.

It is believed that the claims are clear, definite and have full antecedent basis.

This rejection is believed to be overcome, and withdrawal thereof is respectfully requested.

**Conclusion**

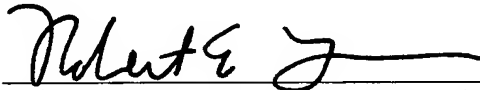
Every issue set forth in the Official Action mailed February 12, 2008 and the Advisory Action mailed April 21, 2008 has been fully addressed.

The objections and rejections are thus believed to have been overcome, obviated or rendered moot, and that no issues remain. The Examiner is accordingly respectfully requested to place the application in condition for allowance and to issue a Notice of Allowability.

The Commissioner is hereby authorized in this, concurrent, and future replies, to charge payment or credit any overpayment to Deposit Account No. 25-0120 for any additional fees required under 37 C.F.R. § 1.16 or under 37 C.F.R. § 1.17.

Respectfully submitted,

YOUNG & THOMPSON



---

Robert E. Goozner, Reg. No. 42,593  
209 Madison Street  
Suite 500  
Alexandria, VA 22314  
Telephone (703) 521-2297  
Telefax (703) 685-0573  
(703) 979-4709

REG/mjr

**APPENDIX:**

The Appendix includes the following item(s):

- A portion of Chapter 7 of *Piezoelectric Ceramics* by B. Jaffe, in which the relationship of PZT, phase structure and morphology is discussed.

# NON-METALLIC SOLIDS

A SERIES OF MONOGRAPHS

*Editors*

J. P. ROBERTS

*The Houldsworth School of Applied Science  
The University, Leeds, England*

P. POPPER

*British Ceramic Research Association  
Stoke-on-Trent, England*

Volume 1. P. W. McMillan: GLASS-CERAMICS. 1964

Volume 2. H. Rawson: INORGANIC GLASS-FORMING SYSTEMS.  
1967

Volume 3. B. Jaffe, W. R. Cook Jr. and H. Jaffe: PIEZOELECTRIC  
CERAMICS. 1971

# PIEZOELECTRIC CERAMICS

*by*

BERNARD JAFFE

*Vernitron Corp.*

*Bedford, Ohio, U.S.A. 44146*

*and*

WILLIAM R. COOK, JR. AND HANS JAFFE

*Gould Inc.*

*Cleveland, Ohio, U.S.A. 44108*

1971



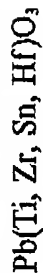
ACADEMIC PRESS  
LONDON and NEW YORK

SIEMENS AKTIENGESELLSCHAFT  
Verlags- und Patentabteilung 3984a

49. Shirane, G., Sawaguchi, E. and Takagi, Y. (1951). *Phys. Rev.* **84**, 476-81.
50. Sawaguchi, E. (1953). *J. Phys. Soc. Japan* **8**, 615-29.
51. Roberts, S. (1951). *Phys. Rev.* **83**, 1078.
52. Jona, F., Shirane, G., Mazzi, F. and Pepinsky, R. (1957). *Phys. Rev.* **105**, 849-66.
53. Shirane, G. and Hoshino, S. (1954). *Acta Cryst.* **7**, 202-10.
54. Berlincourt, D. and Krueger, H. H. A. (1959), personal communication.
55. Gubkin, A. N. and Sorokin, V. S. (1960). *Izv. Akad. Nauk SSSR, Ser. Fiz.* **24**, 246-52.
56. Shirane, G. and Hoshino, S. (1952). *Phys. Rev.* **86**, 248-9.
57. Krainik, N. N. (1958). *Zh. Tekhn. Fiz.* **28**, 525-35; *Soviet Phys. Tech. Phys.* **3**, 498-502.
58. Shirane, G. and Suzuki, K. (1952). *J. Phys. Soc. Japan* **7**, 333.
59. Marutake, M. and Ikeda, T. (1955). *J. Phys. Soc. Japan* **10**, 424-8.
60. Shirane, G. and Pepinsky, R. (1953). *Phys. Rev.* **91**, 812-15.
61. Berlincourt, D., Krueger, H. H. A. and Jaffe, B. (1964). *Phys. Chem. Solids* **25**, 859-74.
62. Krainik, N. N. (1960). *Fiz. Tverd. Tela* **2**, 865-80; *Soviet Phys. Solid State* **2**, 833-7.
63. Onchi, H., Nagano, K. and Hayakawa, S. (1965). *J. Am. Ceram. Soc.* **48**, 630-5.
64. Smolenskii, G. A. and Agranovskaya, A. I. (1964). *Dokl. Akad. Nauk SSSR* **97**, 237-8.
65. Jaffe, B., Roth, R. S. and Marzullo, S. (1955). *J. Res. Nat. Bur. Std.* **55**, 239-54.
66. Smolenskii, G. A., Agranovskaya, A. I., Kalinina, A. M. and Fedotova, T. M. (1955). *Zh. Tekhn. Fiz.* **25**, 2134-42.
67. Sugawara, F., Syono, Y. and Akimoto, S. (1968). *Mat. Res. Bull.* **3**, 529-32.
68. Wilcox, D. L. and Cook, R. L. (1963). *J. Am. Ceram. Soc.* **46**, 343-8.
69. Gavril'yachenko, V. G., Spinko, R. I., Martynenko, M. A., and Fesenko, E. G. (1970). *Fiz. Tverd. Tela* **12**, 1532-7; *Soviet Phys.—solid state* **12**, 1203-7.
70. Remeika, J. P., and Glass, A. M. (1970). *Mat. Res. Bull.* **5**, 37-45.

## Chapter 7

## SOLID SOLUTIONS OF



Ceramics of piezoelectric  $\text{Pb}(\text{Ti, Zr})\text{O}_3$  solid solutions have achieved wide usage in recent years because of their superior properties. The ferroelectric nature of these materials was established in Japan early in the 1950's. Shortly afterward, their unusual piezoelectric properties were brought to light at the National Bureau of Standards. Since then, intensive work has been done on these materials in the authors' laboratory. Unreferenced material in this chapter may be presumed to have originated there. We are indebted to our colleagues, particularly Mr D. Berlincourt, Mr H. Krueger, Mr F. Kulcsar, and Dr R. Gerson (now at Department of Physics, University of Missouri School of Mines and Metallurgy) for many of the results given.

A.  $\text{Pb}(\text{Ti, Zr})\text{O}_3$  SOLID SOLUTIONS1.  $\text{Pb}(\text{Ti, Zr})\text{O}_3$  Phase Diagram

A current version of the phase diagram is shown in Fig. 7.1. Substitution of  $\text{Zr}^{4+}$  for  $\text{Ti}^{4+}$  in  $\text{PbTiO}_3$  reduces the tetragonal distortion (Fig. 7.2) and ultimately causes the appearance of another ferroelectric phase of rhombohedral  $R3m$  symmetry.<sup>1, 2</sup> The boundary between tetragonal and rhombohedral forms is nearly independent of temperature (morphotropic).<sup>3</sup> Still more  $\text{Zr}^{4+}$  causes the appearance of the orthorhombic antiferroelectric  $\text{PbZrO}_3$  phase<sup>4</sup> with a small field of stability of a tetragonal antiferroelectric phase near the Curie point.<sup>5</sup> Its exact extent is rather impurity sensitive, and has not yet been determined with ultra-pure compositions. Low-hafnium zirconia was used in the determination of Fig. 7.1. The orthorhombic and

\* This is used to denote an abrupt structural change within a solid solution with variation in composition as explained in ref. 14.

tetragonal antiferroelectric phases have multiple cells, indicated by superstructure lines.

The rhombohedral ferroelectric phase actually divides into two phases,<sup>4</sup> as shown by measurement of both electrical properties (Fig. 7.3) and thermal expansion (Fig. 7.4). Although

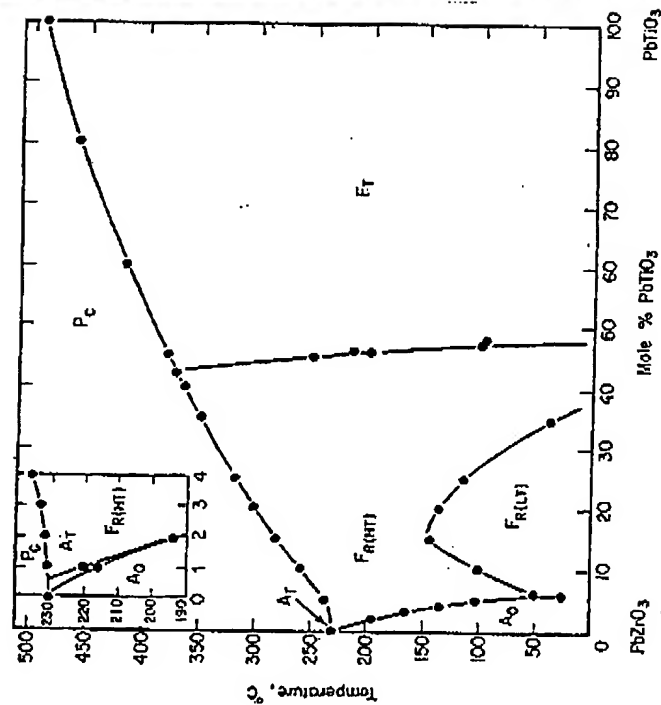


Fig. 7.1.  $\text{PbTiO}_3$ - $\text{PbZrO}_3$  sub-solidus phase diagram.

both appear to be simple cell rhombohedral by X-ray diffraction, neutron diffraction studies indicate a multiple rhombohedral cell for the low temperature phase.<sup>7</sup>

In the later discussion the morphotropic tetragonal-rhombohedral phase boundary will be referred to a specific composition. It is required that this be a two-phase zone and this has been experimentally confirmed. The "phase boundary" is considered as that composition where the two phases are present in equal quantity.

Savaguchi's comprehensive paper<sup>8</sup> explained the stabilities of the phases and analyzed the field-enforced double hysteresis

loops of compositions near  $\text{PbZrO}_3$  on the basis of compositional difference of the free energies of the various phases (Fig. 6.7.)

In the system  $\text{PbO-TiO}_2\text{-ZrO}_2$  (Fig. 7.5), the only stable intermediate phases are probably the  $\text{Pb}(\text{Ti}, \text{Zr})\text{O}_3$  solid solution and  $\text{ZrTiO}_4$ .<sup>8,9</sup> Lead oxide content in the  $\text{Pb}(\text{Ti}, \text{Zr})\text{O}_3$  solid

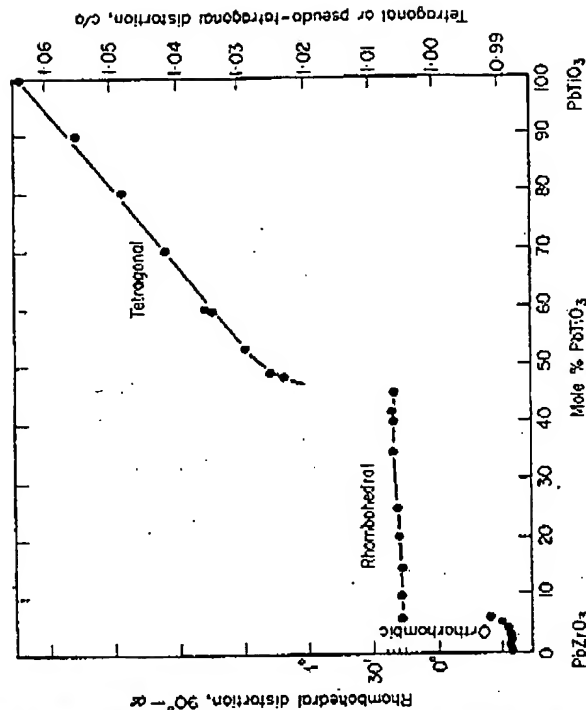


Fig. 7.2. Unit cell distortion at room temperature for the  $\text{PbTiO}_3$ - $\text{PbZrO}_3$  system.

solution can vary between 48 and 51 mole %  $\text{PbO}$ .<sup>10</sup> Higher values previously reported for  $\text{PbO}$  excess and deficiency limits in the solid solution<sup>8,9</sup> are probably due to a combination of volatility of  $\text{PbO}$  and the difficulty of detecting a second phase in a mixture. Limited solid solution exists in all three end members. The recently reported  $\text{PbTi}_3\text{O}_7$ <sup>10a</sup> may also be stable.

$\text{PbTiO}_3$  melts congruently, and  $\text{PbZrO}_3$  melts incongruently. Melting of the solid solution changes from congruent to incongruent at  $\sim 1340^\circ$  (Fig. 7.6.)

The solid state reaction usually begins by formation of a highly tetragonal lead titanate solid solution into which the remaining  $\text{PbO}$  and  $\text{ZrO}_2$  react to form  $\text{Pb}(\text{Ti}, \text{Zr})\text{O}_3$ . Infrequent exceptions have been noted, however, in which the course of the